

# Efficient, long-range correlation from occupied wavefunctions only

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Via continuum mechanics [PRL 103,086401] with Random Phase Approximation (dRPA) screening, we develop a numerically efficient general-geometry electronic exchange-correlation energy functional. It gives correct asymptotic power laws for dispersion interactions between insulators or metals. As a numerical example we obtain the full binding energy curves  $\bar{\epsilon}(D)$  for parallel metal slabs of small but finite thickness: at all separations  $D$  our  $\bar{\epsilon}(D)$  agrees better with full dRPA correlation calculations than does the Local Density Approximation, while being much more efficient than full dRPA correlation.

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An increasing body of work[1–4] has demonstrated that the correlation energy  $E_c^{\text{dRPA}}$  in the direct Random-Phase Approximation (dRPA) is highly accurate for energy differences in many and varied electronic systems, at least in cases where orbital self interaction is not an issue. dRPA binding properties for a wide variety of bulk materials[2] are typically more accurate than those from the local density approximation (LDA), especially for dispersion (van der Waals, vdW) bound systems[3]. For the vdW attractive potential, which is totally neglected in the LDA, the dRPA proves to be versatile, predicting unusual vdW coefficients[5] and power laws[6] in agreement with quantum Monte Carlo results[7].

$E_c^{\text{dRPA}}$  is typically obtained in three steps: i) The bare response  $\hat{\chi}_0$  is obtained from occupied and unoccupied groundstate wavefunctions. This is typically the numerical bottleneck. Recent developments[8] attempt to bypass unoccupied states but can encounter problems for metallic systems. ii) The interacting response is calculated through the dRPA as  $\hat{\chi}_\lambda(\omega) = \hat{\chi}_0(\omega) + \lambda \hat{\chi}_0(\omega) \hat{v} \hat{\chi}_\lambda(\omega)$  where  $\hat{v}$  is the Coulomb potential  $|\mathbf{r} - \mathbf{r}'|^{-1}$ . iii) Finally the correlation energy is calculated via integration on the imaginary frequency axis through the Adiabatic Connection and Fluctuation Dissipation Theorem (ACFD) approach

$$E_c^{\text{dRPA}} = - \int_0^\infty \frac{d\sigma}{2\pi} \text{Tr} \left[ \log \{ \hat{1} + \hat{A}(i\sigma) \} - \hat{A}(i\sigma) \right] \quad (1)$$

where  $\hat{A}(\omega) = -\hat{v}^{1/2} \hat{\chi}_0(\omega) \hat{v}^{1/2}$  is an Hermitian operator[9].

Other efficient van der Waals (vdW) functionals[10, 11] give good results for many systems. However they represent  $E_c^{\text{vdW}}$  in an additive two-point approximation that is either obtained semi-empirically[10] or derived[11] by solving the dynamical screening problem (1) perturbatively. As a result, these functionals miss non-pairwise-additive vdW energy contributions that can be substantial in highly polarizable, highly anisotropic systems[5, 6], including low-dimensional metals. Very large, anisotropic molecules and metallic and graphitic surface physics (e.g. binding of graphite on metal sur-

faces) are two classes of systems where standard methods are inaccurate[12] and dRPA is intractable.

Here we solve equation (1) accurately thus avoiding the pairwise additive approximation, but we use the continuum mechanics of Tokatly, Tao, Gao and Vignale[13, 14] to approximate  $\hat{\chi}_0$  in a numerically efficient manner. Their linearized continuum mechanics (CM) scheme [14] uses the continuum fluid displacement  $\mathbf{u}$ , which is related to the density perturbation  $n^1$  by[14, 15]

$$n^1(\mathbf{r}, t) = -\partial_\mu [n^0(\mathbf{r}) u_\mu(\mathbf{r}, t)]. \quad (2)$$

For a small change to the Kohn-Sham (KS) potential  $V^1(\mathbf{r}, t)$  CM theory approximates  $\mathbf{u}$  through the following hydrodynamic-like equation (from equations 3, 4 and 14-16 of [14])

$$\partial_{tt} u_\mu(\mathbf{r}, t) = \frac{-\Phi_{\mu\nu}^0 u_\nu(\mathbf{r}, t) + F_\mu^0(\mathbf{r}, t)}{n^0(\mathbf{r})} - \partial_\mu V^1(\mathbf{r}, t) \quad (3)$$

where  $n^0(\mathbf{r})$ ,  $\Phi_{\mu\nu}^0 = -n^0(\mathbf{r})[\partial_{\mu\nu} V^{\text{KS}}(\mathbf{r})]$  and  $F_\mu^0(\mathbf{r}, t)$  depend on groundstate properties of the system.

The force  $F_\mu^0$  is defined in equation 14 of [14]. Careful manipulation of equation 14 allows us to write it as  $F_\mu^0 = -\hat{K}_{\mu\nu} u_\nu(\mathbf{r}, t)$ . Here  $\hat{K}$  is a tensor, Hermitian ( $\hat{K}_{\mu\nu} = \hat{K}_{\nu\mu}^\dagger$ ) operator defined by

$$\hat{K}_{\mu\nu} = \hat{K}_{\mu\nu}^{(T)} - \frac{1}{4} \hat{K}_{\mu\nu}^{(n)} \quad (4)$$

$$\hat{K}_{\mu\nu}^{(T)} = \partial_\alpha \bar{T}_{\mu\nu}^0 \partial_\alpha + \partial_\nu \bar{T}_{\mu\alpha}^0 \partial_\alpha + \partial_\alpha \bar{T}_{\alpha\nu}^0 \partial_\mu \quad (5)$$

$$\hat{K}_{\mu\nu}^{(n)} = \partial_{\nu\alpha} n^0(\mathbf{r}) \partial_{\alpha\mu}. \quad (6)$$

It involves the electron density and groundstate kinetic stress tensor  $\bar{T}_{\mu\nu}^0 = \Re \sum_i f_i [\partial_\mu \psi_i(\mathbf{r})]^* [\partial_\nu \psi_i(\mathbf{r})] - \frac{[\partial_{\mu\nu} n^0(\mathbf{r})]}{4}$  [16] where the sum is over occupied orbitals.

In the absence of an external potential, equation (3) has time-periodic eigen-solutions defined by the hydrodynamic eigen-equation

$$-\Omega_N^2 n^0 u_{N\mu}(\mathbf{r}) = [\Phi_{\mu\nu}^0 + \hat{K}_{\mu\nu}] u_{N\nu}(\mathbf{r}) \quad (7)$$

where  $N$  labels the sorted eigen-modes,  $\mathbf{u}_N(\mathbf{r})$  is related to an eigen-function of  $\hat{\chi}_0$ ,  $\Omega_N > \Omega_{N-1}$  is related to the

KS excitation energies (exactly in one-electron systems) and  $\int d\mathbf{r} n^0(\mathbf{r}) \mathbf{u}_N^*(\mathbf{r}) \cdot \mathbf{u}_M(\mathbf{r}) = \delta_{NM}$ .

By definition the tensor polarizability  $X_{0\mu\nu}(\mathbf{r}, \mathbf{r}'; \omega)$  is the time-periodic response of the  $\mu$  cartesian component of the polarization  $-n^0(\mathbf{r}) \mathbf{u}_N(\mathbf{r})$  to an external electric field in the  $\nu$  direction, while  $\chi_0$  is the change in density  $n^1(\mathbf{r})$  in response to a small change in the KS potential of form  $V^1(\mathbf{r}; \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}')$ . They can be obtained through equations (2)-(3) and expansion in the eigen-solutions of (7) provides the convenient forms (where in practise  $N$  is summed over the lowest  $N_{\text{Eig}}$  eigen-pairs)

$$X_{0\mu\nu}(\mathbf{r}, \mathbf{r}', i\sigma) = \sum_N F_N(i\sigma) p_{N\mu}^*(\mathbf{r}) p_{N\nu}(\mathbf{r}') \quad (8)$$

$$\chi_0(\mathbf{r}, \mathbf{r}', i\sigma) = - \sum_N F_N(i\sigma) d_N^*(\mathbf{r}) d_N(\mathbf{r}') \quad (9)$$

where  $F_N(i\sigma) = (\Omega_N^2 + \sigma^2)^{-1}$ ,  $\mathbf{p}_N = n^0(\mathbf{r}) \mathbf{u}_N(\mathbf{r})$  and  $d_N(\mathbf{r}) = [-\nabla \cdot \{n^0(\mathbf{r}) \mathbf{u}_N(\mathbf{r})\}] = -[\nabla \cdot \mathbf{p}_N]$ .

For efficient evaluation of the correlation energy, the following important relationships are derived which allow us to evaluate  $E_c$  using integrals over *one space variable* only, reducing calculation time and storage requirements[17]. From (9) the projection of  $\hat{A}$  [see (1)] in reciprocal space can be written in the separable form  $A(\mathbf{q}, \mathbf{q}') = \sum_N F_N(i\sigma) w_N^*(\mathbf{q}) w_N(\mathbf{q}')$  where

$$w_N(\mathbf{q}) = -i\mathbf{q} \sqrt{v(\mathbf{q})} \cdot \int d\mathbf{r} e^{i\mathbf{q} \cdot \mathbf{r}} n^0(\mathbf{r}) \mathbf{u}_N(\mathbf{r}) \quad (10)$$

or  $w_N = \sqrt{v(\mathbf{q})} d_N(\mathbf{q})$  (here  $v(\mathbf{q}) = 4\pi q^{-2}$ ). Setting  $W_{NM} = \int \frac{d\mathbf{q}}{(2\pi)^3} w_N(\mathbf{q}) w_M^*(\mathbf{q})$  allows us to define an  $N_{\text{Eig}} \times N_{\text{Eig}}$  matrix  $\mathbb{B}(i\sigma)$  with elements

$$B_{NM}(i\sigma) = \sqrt{F_N(i\sigma) F_M(i\sigma)} W_{NM} \quad (11)$$

with  $\lim_{N_{\text{Eig}} \rightarrow \infty} \text{Tr}_N[G(\mathbb{B}(i\sigma))] = \text{Tr}_{\mathbf{q}}[G(\hat{A}(i\sigma))]$ [18] for any analytic function  $G$ .

Finally, defining the eigen-values of  $\mathbb{B}(i\sigma)$  to be  $\beta_\kappa(i\sigma)$  we reduce the correlation energy (1) to the form

$$E_c^{\text{CM}} = - \int_0^\infty \frac{d\sigma}{2\pi} \sum_\kappa \{\log[1 + \beta_\kappa(i\sigma)] - \beta_\kappa(i\sigma)\}. \quad (12)$$

In practice we seem only to need a small number  $N_{\text{Eig}}$  of eigen-solutions to converge correlation energies to a sufficiently small error ( $\propto 1/\Omega_{N_{\text{Eig}}}^3$ ) within CM theory[18]. This agrees with other observations (e.g. ref. 8) that calculating  $E_c$  through a diagonalisation of  $\hat{\chi}_0 \hat{v}$  requires few eigenvalues for convergence.

The most trying calculation in this functional method is evaluation of equation (7), as  $\hat{K}$  is a spatially-dependent, differential operator. To overcome this problem we use an auxiliary basis set  $\mathcal{B} \equiv \{\phi_j(\mathbf{r})\}$ , of size  $N_{\text{Bas}}$ , which need not be mutually orthogonal but must be complete in the limit  $N_{\text{Bas}} \rightarrow \infty$ . Choice of this basis

is the only part of his scheme that differs for different geometries or systems: for example, plane waves for periodic systems, gaussians for atoms and molecules. With a given basis set we expand our CM eigen-function (7) as  $u_{N\mu}(\mathbf{r}) = \sum_j a_{N\mu}^j \phi_j(\mathbf{r})$  which we substitute into equation (7). This provides a set of  $3N_{\text{Bas}} \times 3N_{\text{Bas}}$  coupled equations

$$-\Omega_N^2 N_{jk}^0 a_{N\mu}^k = \{\Phi_{jk\mu\nu}^0 + K_{jk\mu\nu}\} a_{N\nu}^k \quad (13)$$

while  $N_{jk}^0 (a_{N\mu}^{j*} a_{M\mu}^k) = \delta_{NM}$  sets the orthogonality.

The non-operator terms in these equations are  $N_{jk}^0 = \int d\mathbf{r} n^0(\mathbf{r}) \phi_j^*(\mathbf{r}) \phi_k(\mathbf{r})$  and  $\Phi_{jk\mu\nu}^0 = - \int d\mathbf{r} [n^0(\mathbf{r}) \partial_{\mu\nu} V^{\text{KS}}(\mathbf{r})] \phi_j^*(\mathbf{r}) \phi_k(\mathbf{r})$ . Separating the final term into  $K_{jk\mu\nu} = \int d\mathbf{r} \phi_j^*(\mathbf{r}) \hat{K}_{\mu\nu}(\mathbf{r}) \phi_k(\mathbf{r}) = K_{jk\mu\nu}^{(T)} - \frac{1}{4} K_{jk\mu\nu}^{(n)}$  and using integration by parts gives

$$K_{jk\mu\nu}^{(T)} = - \int \{ \bar{T}_{\mu\alpha}^0 [\partial_\nu \phi_j^*] [\partial_\alpha \phi_k] + \bar{T}_{\alpha\nu}^0 [\partial_\alpha \phi_j^*] [\partial_\mu \phi_k] + \bar{T}_{\mu\nu}^0 [\nabla \phi_j^*] \cdot [\nabla \phi_k] \} d\mathbf{r} \quad (14)$$

$$K_{jk\mu\nu}^{(n)} = \int n^0 [\partial_\nu \nabla \phi_j^*] \cdot [\partial_\mu \nabla \phi_k] d\mathbf{r} \quad (15)$$

where all terms are functions of  $\mathbf{r}$  and all derivatives can, ideally, be performed *analytically* on the basis functions.

Surprisingly for a hydrodynamic-style approach, CM theory gives the exact bare responses  $\mathbf{X}_0, \chi_0$  to irrotational fields of one- and two( $\uparrow\downarrow$ )-electron systems around their groundstate[13, 18]. This means that our correlation scheme will give the same results as dRPA for the asymptotic vdW interaction between two hydrogen or two helium atoms.

To explore this further we follow [14] in expanding both the KS and CM response to  $O(\omega^{-4})$  leading to the following identities[18]

$$1 = \sum_{ja} h_{jaN}, \quad \Omega_N^2 = \sum_{ja} h_{jaN} \omega_{ja}^2. \quad (16)$$

Here  $h_{jaN} = \frac{2|f_j - f_a| |K_{jaN}|^2}{|\omega_{ja}|}$  where  $\omega_{ja}$  is a KS eigen-energy difference of an occupied orbital  $|j\rangle$  and unoccupied orbital  $|a\rangle$  with occupations  $f_j$  and  $f_a$ , and  $K_{jaN} = \int \langle j | \hat{\mathbf{J}} | a \rangle \cdot \mathbf{u}_N d\mathbf{r}$  is a mode-overlap matrix element of the current (obtained via the current operator  $\hat{\mathbf{J}}$ ). Thus  $\Omega_N \geq \epsilon_L - \epsilon_H$  for isolated systems and  $\Omega_{N\mathbf{q}} \geq \min_{\mathbf{k}} (\epsilon_{L\mathbf{k}+\mathbf{q}} - \epsilon_{H\mathbf{k}})$  for periodic systems where L labels the lowest unoccupied orbital or band and H labels the highest occupied.

One implication of this is that a Kohn-Sham insulator will remain an insulator under CM, in the sense of finite responses (8)-(9) as  $\sigma \rightarrow 0$ . Thus[18] CM theory obeys the well-known vdW laws for insulators with (e.g.) a  $-C_4 D^{-4}$  asymptotic binding for two thin layers. This is a very strong feature of the CM theory, not shared by common approximated ACFD theories [11, 19] where

explicit cutoffs have to be imposed in the tails in order to suppress metallic-like response.

In the opposite limit of a homogeneous electron gas (HEG), CM is analytically soluble, agrees with the true  $\chi_0$  for  $q \ll k_F$ ,  $\omega \gg v_F q$ , and in particular has a “metallic” infinite polarizability,  $\chi_0 \rightarrow \infty$  as  $q$  and  $\omega \rightarrow 0$ . Electron-gas-like (metallic) systems nevertheless pose a difficult test for CM theory because the single-particle-like excitations occurring for  $\omega < v_F q$ , (and thus not accurately described by CM), can make significant contributions to the RPA correlations, mainly at short spatial range (large wavelength).

This inaccuracy can be improved in metallic systems by employing range-separation (RS) such that the short-range physics is treated by a local scheme. This makes *no contribution* to vdW asymptotic physics. A well-studied RS scheme is described in [20]. It involves choosing a  $q_{RS}$  and splitting up the Coulomb potential, with a long-range component  $v^{(q_{RS})}(r) = \text{erf}(q_{RS}r)r^{-1}$ , equivalent to replacing (10) by  $w_N^{(q_{RS})}(\mathbf{q}) = w_N(\mathbf{q})e^{-q^2/(8q_{RS}^2)}$ . We label the corresponding correlation energy  $E_c^{\text{lrCM}(q_{RS})}$ . This has the additional benefit of accelerating convergence.

For  $\hat{\chi}_0$  to be reliably approximated by continuum mechanics without a separate treatment of the low frequencies we must choose  $q_{RS}$  to be *substantially* less than  $k_F$ . Here we use  $q_{RS} = 0.25r_s^{-1} = 0.13k_F$  where  $r_s$  is a global measure of the inter-electron distance. For the jellium slab problems studied below we simply choose  $r_s$  corresponding to the background charge density of each slab, though more general prescriptions exist. The remaining correlation must be included from local approximations so that

$$E_c^{\text{CM}(q_{RS})}[n] = E_c^{\text{lrCM}(q_{RS})} + \int d\mathbf{r} n(\mathbf{r}) \epsilon_c^{\text{LSr}(q_{RS})} \quad (17)$$

where  $\epsilon_c^{\text{LSr}(q_{RS})}$  is the correlation energy per electron of the HEG with a short-ranged interaction, taken from [21].

Ideally we must also implement a range-separation for exchange, but this proves numerically difficult for the slab geometries we investigate. We instead use the ratio of the long-range exchange to total exchange of an HEG  $A_x \approx 1.1q_{RS}r_s/\sqrt{1+(1.1q_{RS}r_s)^2}$  as a prefactor for the exact exchange (EXX)  $E_x^{\text{EXX}} = -\frac{1}{2} \int d\mathbf{r} d\mathbf{r}' |\mathbf{r} - \mathbf{r}'|^{-1} |\sum_n f_n \psi_n^*(\mathbf{r}) \psi_n(\mathbf{r}')|^2$  and make up the remainder with the LDA. Combining this with (17) gives  $E_{xc} = A_x E_x^{\text{EXX}} + (1 - A_x) E_x^{\text{LDA}} + E_c^{\text{CM}(q_{RS})}$ .

As a numerical test of our proposed functional we choose the difficult case of two thin metal slabs described in Refs. 19, 22, and 23. This system is defined by three parameters only: the width of the slabs  $s$ , the inner surface-surface distance  $D$  and the positive background charge electron density  $\rho = 3/(4\pi r_s^3)$ . The total number of electrons per unit area is  $N_s = 2s\rho = \int_{-\infty}^{\infty} n^+(z) dz$ .

We test our method on slab pairs with  $s = 3a_0$ ,  $r_s = 1.25a_0$  and  $s = 5a_0$ ,  $r_s = 2.07a_0$  which have been studied

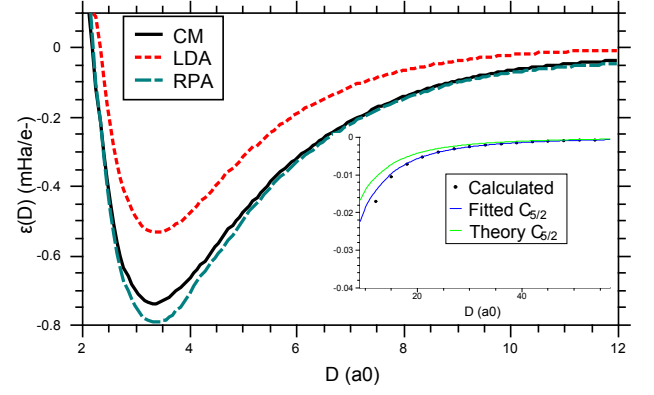


FIG. 1.  $\bar{\epsilon}(D)$  graph for  $r_s = 1.25$ ,  $s = 3$ . RPA data from [23]. Inset data shows the vdW dominated region.

in Ref. 23 and Refs. 19 and 22 respectively. Especially in the first case the LDA and dRPA give significantly different energy curves. We consider the cleavage energy per electron  $\bar{\epsilon}(D) = \epsilon(D) - \epsilon(\infty) = [E_0(D) - E_0(\infty)]/N_s$  as a function of  $D$ . Slabs with  $r_s < 4$  have a defined binding length  $D_0$  where the force is zero. Thus a binding energy  $\epsilon_b = |\bar{\epsilon}(D_0)|$  and an elastic modulus  $C_{zz} = \partial_{DD}\bar{\epsilon}(D_0)$  can also be defined.

In Figure 1 we plot  $\bar{\epsilon}(D)$  versus  $D$  for  $r_s = 1.25$ ,  $s = 3$ . Our method matches the RPA closely for this system. Binding properties for both studied systems are tabulated in Table I and show that the  $r_s = 2.07$ ,  $s = 5$  system is less well-predicted but still much better than the LDA. If instead we set  $q_{RS} = \infty$  the results become much worse for both cases. For widely separated slabs ( $D \gg s$ ) the CM theory correctly and analytically describes coupled two-dimensional plasmons and hence correctly predicts the known asymptotic dRPA form [6]  $\bar{\epsilon}(D \gg s) \approx -0.012562\sqrt{N_s}(D+s)^{-5/2}$ . With  $s = 12.8a_0$  and  $r_s = 2a_0 \dots 6a_0$  we calculate  $C_{5/2}$  numerically within 8% of the theory. By contrast most other efficient vdW functionals would predict an incorrect power law exponent in this limit with  $\bar{\epsilon}(D) \approx -C_4 D^{-4}$ .

In our CM calculations we use auxiliary basis functions  $\phi_{k\mathbf{q}_{\parallel}}(\mathbf{r}) = b_k(z)e^{-i\mathbf{q}_{\parallel} \cdot \mathbf{r}_{\parallel}}$  [18] at  $N_{q_{\parallel}}$   $\mathbf{q}_{\parallel}$  points. All calculations are quite efficient with the slowest step being evaluation of  $W_{NM}$  at  $O(N_{q_{\parallel}} N_{q_z} N_{\text{Bas}}^2)$ . Convergence is reached with  $N_{\text{Bas}} = 42$ ,  $N_{q_{\parallel}} = 55$ ,  $N_{\sigma} = 250$  and

	CM	LDA	dRPA	CM	LDA	dRPA
	$r_s = 1.25, s = 3$			$r_s = 2.07, s = 5$		
$D_0$	3.33	3.38	3.32‡	1.57	1.56	1.62±0.1§
$\epsilon_b$	0.74	0.53	0.79‡	1.78	1.72	1.85±0.1§
$C_{zz}$	0.51	0.45	0.55‡	1.31	1.38	1.32±0.1§

TABLE I. Groundstate properties of two slab systems under different approximations. Energies are in mHa/e⁻ and distance are in Bohr radii. ‡ from Ref. 23, § is guessed from Refs. 19 and 22 taking into account estimated error bars.

$N_{\text{Eig}} \leq 60$ . Our dRPA calculation takes approximately eight times longer than the groundstate LDA calculation. Test runs of full RPA calculations for these systems took hours, compared to minutes for our functional, consistent with  $N_{\text{qll}} = 55$ . We also note that a 10% variation in  $q_{\text{RS}}$  made only a 1% change to  $\epsilon_b$ .

While results for our test systems are not perfect, they show closer agreement with the dRPA than the LDA both in the binding region and for larger  $D$ , with a marked improvement in speed over full dRPA. The vdW dispersive physics is treated accurately and shows excellent agreement with the dRPA in contrast to other methods. The current prescription has a wide scope for refinement both empirically through adjustment of  $q_{\text{RS}}$  and the exchange functional and by introducing better physics, most obviously through improved (semi-local) treatment of low-frequency behaviour which will reduce dependence on the range separation.

Furthermore preliminary tests suggest that metals are a worst-case for CM theory - i.e. that range separation will be much less needed for bound and insulating systems.

In summary, we have derived and developed an *efficient* general-geometry functional with correct long-range correlation energy. Its ability to predict correctly the vdW physics of metallic and insulating systems is a distinct advantage over other efficient vdW functionals. It is currently being implemented for periodic systems, which should enable meaningful energy calculations for (e.g.) vdW bonded nanosystems such as metallic nanotube arrays or graphene on metals. These systems require non-pair-additive high-level computations (e.g. dRPA) with a large unit cell, which is beyond present computational power.

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# Supplementary material for “Efficient, long-range correlation from occupied wavefunctions only”

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## EQUIVALENCE OF TRACES

The calculation of  $E_c$  via  $W$  relies on an equivalence of certain traces. Here we demonstrate that

$$\text{Tr}_{\mathbf{q}}[\log(1 + \hat{A}) - \hat{A}] = \text{Tr}_N[\log(\mathbb{I} + \mathbb{B}) - \mathbb{B}] \quad (1)$$

where  $\hat{A} = -\hat{v}^{\frac{1}{2}}\hat{\chi}_0\hat{v}^{\frac{1}{2}}$ ,  $B_{NM} = \sqrt{F_N(i\sigma)F_M(i\sigma)}W_{NM}$  and  $W_{NM} = \int \frac{d\mathbf{q}}{(2\pi)^3} w_N(\mathbf{q})w_M^*(\mathbf{q})$ .

Let us project  $\hat{A}$  onto Fourier space giving

$$\hat{A}(\mathbf{q}, \mathbf{q}') = \sum_{NM} [\mathbb{F}]_{NM} w_N^*(\mathbf{q})w_M(\mathbf{q}') \quad (2)$$

where  $F_{NM} = F_N(i\sigma)\delta_{NM}$  and we subsequently omit  $\sigma$  for notational brevity. We can then evaluate

$$\begin{aligned} \hat{A}^2(\mathbf{q}, \mathbf{q}') &= \int \frac{d\mathbf{q}_2}{(2\pi)^3} \hat{A}(\mathbf{q}, \mathbf{q}_2) \hat{A}(\mathbf{q}_2, \mathbf{q}') \\ &= \int \frac{d\mathbf{q}_2}{(2\pi)^3} \sum_{N_1 M_1} [\mathbb{F}]_{N_1 M_1} w_{N_1}^*(\mathbf{q})w_{M_1}(\mathbf{q}_2) \\ &\quad \times \sum_{N_2 M_2} [\mathbb{F}]_{N_2 M_2} w_{N_2}^*(\mathbf{q}_2)w_{M_2}(\mathbf{q}') \\ &= \sum_{NM} w_N^*(\mathbf{q})w_M(\mathbf{q}') [\mathbb{F}\mathbb{W}\mathbb{F}]_{NM} \end{aligned} \quad (3)$$

or more generally

$$\hat{A}^p(\mathbf{q}, \mathbf{q}') = - \sum_{NM} w_N^*(\mathbf{q})w_M(\mathbf{q}') [\mathbb{F}\mathbb{W}^p]_{NM} \quad (4)$$

when  $\hat{M}(\mathbf{q}, \mathbf{q}') = \sum_{NM} w_N^*(\mathbf{q})w_M(\mathbf{q}') [\mathbb{M}]_{NM}$ .

Observation of  $\hat{A}^2$  suggests that

$$\hat{A}^p(\mathbf{q}, \mathbf{q}') = \sum_{NM} w_N^*(\mathbf{q})w_M(\mathbf{q}') [(\mathbb{F}\mathbb{W})^{p-1}\mathbb{F}]_{NM}. \quad (5)$$

Premultiplying by  $\hat{A}$  validates this assumption since

$$\begin{aligned} \hat{A}\hat{A}^p(\mathbf{q}, \mathbf{q}') &= \sum_{NM} w_N^*(\mathbf{q})w_M(\mathbf{q}') [\mathbb{F}\mathbb{W}(\mathbb{F}\mathbb{W})^{p-1}\mathbb{F}]_{NM} \\ &= \hat{A}^{p+1}(\mathbf{q}, \mathbf{q}') \end{aligned} \quad (6)$$

and thus (5) is the correct form by induction on  $p$ .

Finally we can take traces (and use permutations under

a trace) to show

$$\begin{aligned} \text{Tr}_{\mathbf{q}}[\hat{A}] &= \sum_N \int \frac{d\mathbf{q}}{(2\pi)^3} w_N^*(\mathbf{q})w_N(\mathbf{q}) [\mathbb{F}]_{NN} \\ &= \sum_N W_{NN} [\mathbb{F}]_{NN} \\ &= \text{Tr}_N [\mathbb{F}\mathbb{W}] = \text{Tr}_N [\mathbb{B}] \end{aligned} \quad (7)$$

$$\begin{aligned} \text{Tr}_{\mathbf{q}}[\hat{A}^p] &= \sum_{NM} \int \frac{d\mathbf{q}}{(2\pi)^3} w_N^*(\mathbf{q})w_M(\mathbf{q}) [(\mathbb{F}\mathbb{W})^{p-1}\mathbb{F}]_{NM} \\ &= \sum_{NM} W_{MN} [(\mathbb{F}\mathbb{W})^{p-1}\mathbb{F}]_{NM} \\ &= \text{Tr}_N [(\mathbb{F}\mathbb{W})^p] = \text{Tr}_N [\mathbb{B}^p] \end{aligned} \quad (8)$$

where  $\mathbb{B} = \sqrt{\mathbb{F}\mathbb{W}}\sqrt{\mathbb{F}}$  and  $B_{NM} = \sqrt{F_N(i\sigma)F_M(i\sigma)}W_{NM}$ . Finally it follows from Taylor expansion of an analytic function  $G(\hat{A}) = \sum_p g_p \hat{A}^p$  that

$$\text{Tr}_{\mathbf{q}}[G(\hat{A})] = \sum_p g_p \text{Tr}_{\mathbf{q}}[\hat{A}^p] = \text{Tr}_N[G(\mathbb{B})]. \quad (9)$$

Setting  $G(x) = \log(1+x) - x$  gives the equivalence.

## EIGENVALUE CONVERGENCE

In the main work we mention the rapid convergence with respect to the CM eigen-pairs. While this is somewhat system specific some general considerations make it likely that fewer eigen-pairs than Kohn-Sham (KS) transition modes will be required to obtain the same convergence in most systems.

For any given KS problem we can expand solutions in a finite basis set (eg. a real space grid, planewaves, Gaussians etc.) of size  $N_{\text{Bas}}$  which can be made as large as we like. The KS equations thus have  $N_{\text{Bas}}$  solutions of which  $N_{\text{Occ}}$  may be considered occupied and  $N_{\text{Bas}} - N_{\text{Occ}}$  are unoccupied. Since  $\hat{\chi}_0$  involves a sum over occupied and unoccupied states, calculations involving  $\hat{\chi}$  have a leading  $O(N_{\text{Occ}}(N_{\text{Bas}} - N_{\text{Occ}})) \approx O(N_{\text{Occ}}N_{\text{Bas}})$  if we assume  $N_{\text{Occ}} \ll N_{\text{Bas}}$ .

By contrast the tensor CM equations have  $N_{\text{Eig}} = 3N_{\text{Bas}}$  solutions. Projection of  $\hat{\chi}_0$  involves a sum over all solutions and takes  $O(3N_{\text{Bas}})$ . Combining this with the sum rules [see (16) of main text] means that the CM eigen-frequencies will be distributed more sparsely than the KS-transitions (except in one and  $\uparrow\downarrow$  two electron systems).

In the specific case of a periodic system this has a very notable effect on integration over the Brillouin zone. Here a natural basis set for both the LDA and CM is  $e^{i(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}}$  where  $\mathbf{q}$  lies within the Brillouin zone and  $\mathbf{G}$  is a reciprocal lattice vector. It is thus sufficient to project  $\hat{\chi}_0$  onto  $\mathbf{q}$ ,  $\mathbf{G}$  and  $\mathbf{G}'$ . Following the standard notation we use  $\mathbf{k}$  rather than  $\mathbf{q}$  for the groundstate sampling.

In a full KS-dRPA calculation, projection of  $\hat{\chi}_0$  is  $O(N_{\text{Occ band}} N_{\text{Band}} N_{\mathbf{k}}^2 N_{\mathbf{G}}^2)$  where  $N_{\text{Occ band}}$  is the number of occupied bands and  $N_{\text{Band}}$  is the total number of unoccupied bands.  $N_{\mathbf{k}}$  is the number of points sampled in the Brillouin zone and must be counted twice: once for each projection onto  $\mathbf{q}$  and once to cover all transitions from  $\mathbf{k}$  to  $\mathbf{k} + \mathbf{q}$  (see e.g. section III of ref. 1 for a more comprehensive discussion). By contrast the same projection in CM is  $O(N_{\text{Eig}} N_{\mathbf{k}} N_{\mathbf{G}}^2)$  as all  $\mathbf{k}$  to  $\mathbf{k} + \mathbf{q}$  transitions are treated collectively in a single  $\mathbf{u}_{N\mathbf{q}}$  eigenmode.

This relationship can also be observed if we solve for  $\hat{\chi}$  by direct perturbation[2] instead of the diagonalization method used here. In a full KS calculation we need to perturb each of the  $N_{\text{Occ}}$  orbitals to obtain  $\hat{\chi}$ . In the CM we need only to solve for the three-dimensional quantity  $\mathbf{u}$ . In the periodic bulk case  $N_{\text{Occ}} = N_{\mathbf{k}} N_{\text{Occ band}}$ .

With a similar energy cutoff for both the KS and CM solutions so that  $N_{\text{Eig}} \approx N_{\text{Band}}$ , a planewave-based evaluation of the CM-dRPA will run at least  $O(N_{\mathbf{k}})$  times quicker than a full dRPA calculation. In an efficient dRPA code in an insulating system,  $N_{\mathbf{k}}$  is typically found to be converged with between 200 and 2000 points[1] divided by the number of symmetries.

Under the dRPA, the truncation error caused by using only a finite number of transitions, is  $O(1/\omega_{amjm}^3)$  where  $\omega_{amjm} = \epsilon_{a_m} - \epsilon_{j_m}$  (where  $a_m$  is an occupied KS state and  $j_m$  is unoccupied) is the largest transition frequency included in the sum. The equivalent error for the CM is  $O(1/\Omega_{N_m})$  where  $\Omega_{N_m}$  is the transition frequency of the highest included mode.

We must note, however, that there are a variety of ways to calculate  $E_c$ . For example we can bypass the projection of  $\hat{\chi}_0$ , as we do in the method presented in the manuscript, by using  $\mathbb{W}$ . For periodic bulks this is an  $O(N_{\mathbf{k}} N_{\text{Bas}}^2 N_{\mathbf{G}})$  calculation under the CM with  $O(N_{\text{Bas}}^2)$  storage and  $O(N_{\text{Bas}}^3)$  diagonalisation. In the full KS it would be  $O(N_{\mathbf{k}}^3 N_{\text{Occ}}^2 N_{\text{Bas}}^2 N_{\mathbf{G}})$  with  $O(N_{\mathbf{k}}^2 N_{\text{Occ}}^2 N_{\text{Bas}}^2)$  storage and  $O(N_{\mathbf{k}}^3 N_{\text{Occ}}^3 N_{\text{Bas}}^3)$  diagonalisation which is infeasible for most bulk systems.

As a general rule, projection of  $\hat{\chi}_0$  onto space will be more efficient than using  $\mathbb{W}$  for a true KS bulk or periodic system. Calculation of  $\mathbb{W}$  will be quicker for the CM in almost all geometries and will be quicker than evaluation of  $\mathbb{W}$  for the full KS system in all but the smallest systems.

In summary, for systems with more than a handful of occupied orbitals, the CM is all but guaranteed to converge faster than the full KS under the same method for calculating  $E_c$ . Furthermore it may offer the potential

to use alternate methods that are faster still (such as using  $\mathbb{W}$  for correlation energies) which may be infeasible or much slower in the full KS system.

## EXACTNESS IN ONE-ELECTRON SYSTEMS

Equations (41), (45-46) of Ref. 3 provide a proof that the continuum mechanics approach is exact for one-electron (or two-electron with equal spin up and down) systems. This follows from the equivalence of CM to Madelung dynamics which are equivalent to the Schrödinger equation for one-orbital systems in irrotational fields[4]. We will provide a direct proof from the Schrödinger equation elsewhere.

## NON-CONTRIBUTING GAUGE MODES

It is possible to find solutions  $\mathbf{u}_N$  of the CM equations where  $\nabla \cdot n^0 \mathbf{u}_N = 0$ . For example, in the 2DEG systems examined modes of form  $\mathbf{u}_N = \frac{c}{n^0(z)} \mathbf{q}_{\perp} e^{i\mathbf{q}_{\parallel} \cdot \mathbf{r}}$  where  $\mathbf{q}_{\perp} = \mathbf{q}_{\parallel} \times \hat{\mathbf{z}}$  have this property.

For such modes (labelled  $N^*$ ) the weights of the sum rules [equation (16) of main text] become  $h_{jaN^*} = 0$ . While a full proof is beyond the scope of this supplement it follows from the exactness of the CM response to  $O(\omega^{-4})$  and the near-completeness over all occupied/unoccupied pairs  $ja$  of  $\langle j | \hat{\mathbf{J}} | a \rangle$  for vectors with non-zero gradients. Thus  $\sum_{ja} h_{jaN^*} = 0$  and  $\Omega_{N^*}^2 = \sum_{ja} h_{jaN^*} \omega_{ja}^2 = 0$ . One consequence of this is that any CM eigen-mode of a KS-insulating system which has  $\Omega_{N^*} = 0$  must also have  $\nabla \cdot n^0 \mathbf{u}_{N^*} = 0$ .

Such modes will either be suppressed by the boundary conditions of the problem, or not contribute to the correlation energy of the system and may thus be discarded. More precisely they do not contribute to the linear response  $\hat{\chi}_0$  at all and contribute only a shift of gauge to the tensor response  $\mathbf{X}_0$ . The former identity can be seen directly as only  $\nabla \cdot n^0 \mathbf{u}_N$  appears in the expression [equation (9) of main text] for  $\hat{\chi}_0$  while the latter comes indirectly from integration by parts over the Coulomb tensor.

## INTERACTING INSULATOR

We show in the main work that a KS insulating system (at a bare response level) will remain insulating under the CM. More formally this means that both the KS system and the CM approximation thereto, obey

$$|\hat{\chi}_{0\mathbf{q}}(\mathbf{G}, \mathbf{G}'; i\sigma)| \leq \frac{Y}{W^2 + \sigma^2} \quad (10)$$

where  $Y$  and  $W$  are undetermined but non-zero constants. In the CM case this follows from  $\mathbf{p}_N(\mathbf{r}) =$

$n^0(\mathbf{r})\mathbf{u}_N(\mathbf{r}) < \infty$ ,  $\mathbf{p}_N(\mathbf{r} \rightarrow \infty) = 0$  and  $\Omega_{N\mathbf{q}} > 0$  and the asymptote  $\lim_{\sigma \rightarrow \infty} |\hat{\chi}_{0\mathbf{q}}| = C/\sigma^2$ .

Using the  $\mathbf{p}_N$  and  $\mathbf{p}_M$  matrix representation of the RPA-interacting response  $\hat{\chi}_\lambda$  it is possible to show

$$\hat{\chi}_{\lambda\mathbf{q}}(\mathbf{G}, \mathbf{G}'; i\sigma) = \sum_H \sum_{NM} \frac{X_{HNM} \mathbf{p}_{N\mathbf{q}}^*(\mathbf{G}) \mathbf{p}_{M\mathbf{q}}(\mathbf{G}')}{\Omega_{\lambda H\mathbf{q}}^2 + \sigma^2} \quad (11)$$

where  $\Omega_{\lambda H\mathbf{q}}^2$  is an eigenvalue of the matrix  $\mathbb{D}_{\lambda\mathbf{q}} = \text{diag}[\Omega_{N\mathbf{q}}^2] + \lambda \mathbb{W}_{\mathbf{q}}$ . Provided  $\mathbb{D}_{\lambda\mathbf{q}}$  does not have any zero eigenvalues (this would be a strongly correlated metallisation and is extremely rare) it is obvious that

$$|\hat{\chi}_{\lambda\mathbf{q}}(\mathbf{G}, \mathbf{G}'; i\sigma)| \leq \frac{Y'}{W'^2 + \sigma^2} \quad (12)$$

for some finite  $Y'$  and  $W'$ .

### THIN INSULATORS HAVE $D^{-4}$ POWER LAWS

If we have two well-separated electronic systems at a distance  $D$  with no electronic overlap between them, we can write the dRPA dispersion energy in Lifshitz-like form:

$$\begin{aligned} U^{\text{vdW}}(D) &= E_c(D) - E_c(\infty) \\ &= \int_0^\infty \frac{d\sigma}{\pi} T(\sigma) \end{aligned} \quad (13)$$

$$T(\sigma) = \text{Tr} \left[ \log(1 - \hat{\chi}_{1A} \hat{V}_{AB} \hat{\chi}_{1B} \hat{V}_{BA}) \right] \quad (14)$$

where  $\hat{\chi}_{1A/B}$  is the interacting response of system A/B in isolation defined in the dRPA as

$$\hat{\chi}_{1A/B} = \hat{\chi}_{0A/B} + \chi_{0A/B} \hat{v} \chi_{1A/B}. \quad (15)$$

$\hat{V}_{AB} = \hat{V}_{BA}$  is the Coulomb potential between electrons in different systems *only* and can be considered a function of  $\mathbf{r}_A, \mathbf{r}_B$  and  $D$  where  $\mathbf{r}_{A/B}$  is a position in system A/B. Equations (13)-(14) follow from analysis of Feynmann ring diagrams (see [5] for a similar analysis) or from the ACFD where the intra-system Coulomb interaction  $\hat{v}$  and  $\hat{V}_{AB}$  are switched on separately.

Let us define a system composed of two periodic slabs localised in  $z$  (ie. where we can define a length  $s$  such that  $\chi_{1A/B\mathbf{q}}(z, z')$  is negligible for  $|z| \geq s/2$  or  $|z'| \geq s/2$ ) and centered such that  $\int z dz n_{A/B}^0(z) = 0$ . In such a system it can be shown[6] that, in the limit  $D \gg s$  and  $D \rightarrow \infty$ , (14) is equivalent to

$$T(\sigma) \approx \int_{\text{BZ}} d\mathbf{q}_\parallel \left[ \log(1 - \bar{X}_{1A\mathbf{q}_\parallel} \bar{X}_{1B\mathbf{q}_\parallel} q_\parallel^2 e^{-2q_\parallel D}) \right]. \quad (16)$$

Here  $\bar{X}_{1A/B\mathbf{q}_\parallel}(i\sigma) = \int dz \int dz' (\hat{\mathbf{q}}_\parallel - i\hat{z}) \cdot \mathbf{X}_{1A/B\mathbf{q}_\parallel}(\mathbf{0}, z, \mathbf{0}, z'; i\sigma) \cdot (\hat{\mathbf{q}}_\parallel + i\hat{z})$  where we project the tensor response  $\mathbf{X}_{1A/B\mathbf{q}}(\mathbf{G}_\parallel, z, \mathbf{G}'_\parallel, z'; i\sigma)$  in reciprocal space in the  $xy$ -plane and real space in  $z$ .

In an insulating system we have shown that  $|\hat{\chi}_{1\mathbf{q}}(i\sigma)| \leq \frac{Y}{W^2 + \sigma^2}$  and thus  $|\bar{X}_{1\mathbf{q}}(i\sigma)| \leq \frac{Y'}{W'^2 + \sigma^2}$  where  $Y$  and thus  $Y'$  is finite. Since  $W$  is finite there is a well-defined upper bound for insulating systems and we need not worry about singularities in  $\bar{X}_{1A/B\mathbf{q}}$  we can set  $x = q_\parallel D$  and make a series expansion in  $D^{-1}$  such that

$$\begin{aligned} T(\sigma) &\approx \frac{K}{D^2} \int_0^{k_F D} 2\pi x dx \log[1 - \bar{X}_{1A0} \bar{X}_{1B0} \frac{x^2}{D^2} e^{-x}] \\ &\approx -\frac{K'}{D^4} \bar{X}_{1A0} \bar{X}_{1B0} + O(D^{-6}). \end{aligned} \quad (17)$$

Inserting this into (16) we find

$$U^{\text{vdW}}(D) = -\frac{K'}{D^4} \int_0^\infty \frac{d\sigma}{\pi} \bar{X}_{1A0}(i\sigma) \bar{X}_{1B0}(i\sigma) \quad (18)$$

$$= -\frac{C_4}{D^4}. \quad (19)$$

Ergo two insulating slabs have a  $D^{-4}$  power law in CM theory as in full dRPA. This conclusion applies only for insulators: for thin metals the  $\sigma$  integration diverges.

### TWO-SLAB GEOMETRY

Let us define our two-slab metal problem to have a background charge  $n^+(z) = \rho[H(\frac{s}{2} - |z - L|) + H(\frac{s}{2} - |z + L|)]$  where  $L = \frac{(D+s)}{2}$  and  $H(x) = 1 \forall x \geq 0, 0$  otherwise. This defines two jellium slabs of width  $s$ , surface-to-surface distance  $D$  and background charge per unit area  $\rho = 3/(4\pi r_s^3)$ . The total number of electrons per unit area is set to  $N_s = 2s\rho = \int_{-\infty}^\infty n^+(z)$ .

The partial isotropy means  $V^{\text{KS}}(\mathbf{r}) \equiv V^{\text{KS}}(z)$  and the KS wavefunctions take the form

$$\psi_{n\mathbf{k}_\parallel}(\mathbf{r}) = p_n(z) e^{-i\mathbf{k}_\parallel \cdot \mathbf{r}_\parallel} \quad (20)$$

where  $\int dz p_n^*(z) p_m(z) = (2\pi)^{-2} \delta_{nm}$ . The KS energies are  $\epsilon_{n\mathbf{k}_\parallel} = \epsilon_{n0} + \frac{1}{2} |\mathbf{k}_\parallel|^2$  with occupation  $f_n = 2 \max(\epsilon_F - \epsilon_{n0}, 0)$ . The density and kinetic pressure tensor are thus

$$n^0(z) = \sum_n f_n |p_n(z)|^2 \quad (21)$$

$$\bar{\mathbf{T}}^0(z) = t^{0\parallel}(z) [\hat{\mathbf{x}} \otimes \hat{\mathbf{x}} + \hat{\mathbf{y}} \otimes \hat{\mathbf{y}}] + t^{0z}(z) \hat{z} \otimes \hat{z} \quad (22)$$

where  $t^{0\parallel}(z) = \sum_n f_n \frac{\epsilon_F - \epsilon_{n0}}{2} |p_n(z)|^2$  and  $t^{0z}(z) = \sum_n f_n |\partial_z p_n(z)|^2 - \frac{1}{4} \partial_{zz} n^0(z)$ .

For the present slab problem we choose auxiliary basis functions of the form  $\phi_{k\mathbf{q}_\parallel}(\mathbf{r}) = b_k(z) e^{-i\mathbf{q}_\parallel \cdot \mathbf{r}_\parallel}$  where  $b_k(z)$  is either  $e^{in\frac{\pi z}{s}}$  or  $\tanh(k_t z) e^{in\frac{\pi z}{s}}$  where  $k_t$  is a parameter chosen to optimise convergence and  $n$  is an integer. The restriction to integer  $n$  makes this basis set incomplete but inclusion of non-integer  $n$  does not alter results.

We then set

$$\mathbf{u}_{N\mathbf{q}_\parallel} = \sum_k \phi_{k\mathbf{q}_\parallel}(\mathbf{r}) [a_{Nz}^k(q_\parallel) \hat{z} + a_{N\parallel}^k(q_\parallel) \hat{\mathbf{q}}_\parallel] \quad (23)$$

(the  $\hat{\mathbf{q}}_{\perp} = \mathbf{q}_{\parallel} \times \hat{\mathbf{z}}$  term does not contribute to the correlation energy). Thus the eigen-equations are

$$\Omega_N^2(q_{\parallel}) N_{jk}^0 a_{N\parallel}^k(q_{\parallel}) = K_{jk\parallel\parallel}(q_{\parallel}) a_{N\parallel}^k(q_{\parallel}) + K_{jk\parallel z}(q_{\parallel}) a_{Nz}^k(q_{\parallel}) \quad (24)$$

$$\Omega_N^2(q_{\parallel}) N_{jk}^0 a_{Nz}^k(q_{\parallel}) = [\Phi_{jkzz}^0 + K_{jkzz}(q_{\parallel})] a_{Nz}^k(q_{\parallel}) + K_{jkz\parallel}(q_{\parallel}) a_{N\parallel}^k(q_{\parallel}) \quad (25)$$

which must be solved for each  $q_{\parallel}$ . Normalisation gives  $\sum_{jk} N_{jk}^0 [a_{N\parallel}^{j*} a_{M\parallel}^k + a_{Nz}^{j*} a_{Mz}^k] = (2\pi)^{-2} \delta_{NM}$ .

Here  $N_{jk} = \int dz n^0(z) b_j^*(z) b_k(z)$  and  $\Phi_{jkzz}^0 = \int dz n^0(z) [\partial_{zz} V^{\text{KS}}(z)] b_j^*(z) b_k(z)$  are independent of  $q_{\parallel}$ . The components of  $\mathbb{K}(q_{\parallel})$  take the form

$$-K_{jkzz} = 3\mathcal{F}_{jk}[t^{0z}, 1, 1] + \frac{1}{4}\mathcal{F}_{jk}[n^0, 2, 2] + q_{\parallel}^2 \left( \mathcal{F}_{jk}[t^{0z}, 0, 0] + \frac{1}{4}\mathcal{F}_{jk}[n^0, 1, 1] \right) \quad (26)$$

$$-K_{jk\parallel\parallel} = \mathcal{F}_{jk}[t^{0\parallel}, 1, 1] + \frac{q_{\parallel}^4}{4}\mathcal{F}_{jk}[n^0, 0, 0] + q_{\parallel}^2 \left( 3\mathcal{F}_{jk}[t^{0\parallel}, 0, 0] + \frac{1}{4}\mathcal{F}_{jk}[n^0, 1, 1] \right) \quad (27)$$

$$-K_{jk\parallel z} = (-iq_{\parallel}) \left( \mathcal{F}_{jk}[t^{0z} + t^{0\parallel}, 0, 1] + \frac{1}{4}\mathcal{F}_{jk}[n^0, 1, 2] \right) + \frac{-iq_{\parallel}^3}{4}\mathcal{F}_{jk}[n^0, 0, 1]. \quad (28)$$

where we use the shorthand  $\mathcal{F}_{jk}[f, a, b] = \int f(z) [\partial_{za} b_j^*(z)] [\partial_{zb} b_k(z)] dz$ .

Finally in this basis

$$w_{Nq_{\parallel}}(q_z) = iv^{1/2} (\sqrt{q_{\parallel}^2 + q_z^2}) \times \int dz e^{iq_z z} [q_{\parallel} u_{Nq_{\parallel}} + q_z u_{Nq_{\parallel}z}], \quad (29)$$

$$W_{NM}(q_{\parallel}) = - \int \frac{dq_z}{2\pi} w_{Nq_{\parallel}}^*(q_z) w_{Mq_{\parallel}}(q_z). \quad (30)$$

and

$$E_c^{\text{lrCM}(\mu)} = - \int \frac{d\sigma}{2\pi} \int \frac{2\pi q_{\parallel} dq_{\parallel}}{(2\pi)^2} \text{Tr}[L(\mathbb{B}(q_{\parallel}, i\sigma))] \quad (31)$$

$$B_{NM} = \sqrt{f_N(\sigma) f_M(\sigma)} W_{NM}(q_{\parallel}) \quad (32)$$

where  $L(x) = \log(1+x) - x$ .

In our calculations we use approximately 500-1000 regularly distributed  $z$  points for quadrature (with the number depending on system size). We also use approximately 500  $q_z$  points on a Gauss-Hermite grid (due to the Range-Separation term  $e^{-q^2/(2q_{\text{RS}})^2}$  to calculate  $W_{NM}(q_{\parallel})$ ). This is more than sufficient to represent the chosen basis functions in either space.

To correctly integrate over frequency we require a grid that accurately deals with functions of form  $a/(b^2 + \sigma^2)$  where  $b$  ranges from very small to large. Choosing a regular grid for  $\sigma \ll 1$  and using a Clenshaw-Curtis grid for larger  $\sigma$  seems to work well for these problems.

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- [6] A full proof is beyond the scope of this supplement but can be provided on request. It arises from a cancellation of most terms, in the limit  $D \rightarrow \infty$ , due to the dominance of  $|\mathbf{q}_{\parallel}| = O(1/D)$ . This means that  $e^{-|\mathbf{q}_{\parallel}| + \mathbf{G}_{\parallel}|D} \ll e^{-q_{\parallel}D}$  for  $\mathbf{G}_{\parallel} \neq 0$  and  $\int dz X(z) e^{|\mathbf{q}_{\parallel}|z} \approx \int dz X(z)$ .